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Observation of a highly polarized liquid $^3$He state

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Résumé. — Nous décrivons ici une expérience qui prouve qu'à basse température il est possible de préparer un liquide d'hélium 3 avec une forte polarisation magnétique. Celle-ci est obtenue par fusion rapide d'un solide d'hélium 3, lui-même fortement polarisé. Le temps de relaxation de l'aimantation de ce liquide polarisé est identique au temps de relaxation nucléaire spin-réseau $T_1$ du $^3$He liquide non polarisé : 150 s à 300 mK sous 22 bar.

Abstract. — We describe an experiment which indicates that at low temperature it is possible to prepare a liquid of helium 3 with a large magnetic polarization. The latter is obtained by rapid-melting of highly polarized solid helium 3. The relaxation time of the magnetization of this polarized state is essentially the same as the nuclear spin-lattice relaxation time $T_1$ of the non-polarized liquid $^3$He : 150 s at 300 mK under 22 bar.

B. Castaing and P. Nozières have recently proposed [1] that a highly polarized $^3$He liquid phase could be prepared by a rapid adiabatic melting of a highly polarized solid phase of $^3$He. They have calculated several thermodynamic properties of such a transition and they have predicted, using the Fermi liquid and solid $^3$He properties, that the melting curve $p = f(T)$ would be lowered significantly according to the magnetic polarization of the initial state.

Any polarized liquid thus produced, normally has only a transient existence : the non-polarized liquid state is recovered after a time $T_\tau$ which characterizes the relaxation of the Zeeman energy ; the magnetization decreases down to a value calculated within the framework of Fermi liquid theory and given by

$$M_t \simeq \frac{3}{2} \frac{H}{T_F}$$

where $H$ is the external applied field and $T_F$ the Fermi temperature.

In fact, many relaxation processes could be effective during the melting of the solid, as well as strong relaxations in the cell where the liquid is prepared : for instance, relaxation onto the walls might have played a role and it was not clear whether this simple procedure would work or not, even if there are no objections against the principle of the experiment.

The purpose of the report given below is to give experimental evidence that such a polarized liquid helium phase exists for a time comparable to $T_1$, the relaxation time of bulk liquid helium 3 in equilibrium at the same temperature ; it opens the possibility for performing new experiments to test the properties of Fermi-liquids.

1. Principle of the experiment. — The physical idea which sustains the experiment is the following : in the solid phase, the system behaves for our purposes like a Curie paramagnet. At large enough field and
low enough temperature the polarization can reach a reasonable fraction of the magnetization at saturation:

\[ M = \frac{h}{T} \]

(2)

where \( T \) is the temperature of the solid. This is much larger than the magnetization obtained with liquid helium 3 in the same conditions (see formula (1)), because the latter is a Fermi-liquid. The enhancement factor is of the order of \( T_F/T \).

To prepare the liquid we have adopted the following procedure: prepare a solid at high pressure, cool the sample to low temperature, apply large external field, wait long enough to reach an equilibrium temperature and then melt the solid rapidly to get the transient polarized liquid state. By melting rapidly, we mean in time scale short in comparison with the relaxation processes.

The behaviour of the system is observed by measuring the magnetization at each stage of the melting process.

2. Experimental conditions and procedure.

Figure 1 shows the experimental set up. The \(^3\)He is contained in a cell of constant volume thermally coupled to the mixing chamber of a dilution refrigerator \([2]\). Its inner diameter (1 cm) is equal to the height. The heat is transferred from the cell to the mixing chamber by 100 copper wires (\( \Omega = 0.01 \) cm) soldered to a heat exchanger made of sintered silver plates. The cell is placed at the centre of a superconducting magnet which gives a field \( H_0 = 3.5 \) T.

During an experiment (melting of the polarized solid) three parameters can be recorded:

- the \(^3\)He temperature inside the cell is measured by a carbon resistor, the magnetoresistance of which is calibrated using the nuclear susceptibility of the solid \(^3\)He under the same field (Curie law),
- the magnetization of the \(^3\)He is measured by NMR.

The experiments were performed at 114.8 MHz in 3.54 T with a coherent pulsed NMR spectrometer. The transverse coil is almost in a Helmholtz arrangement; it has a \( Q \) factor of the order of 100. We can obtain pulsed rotating magnetic fields of the order of 1 gauss with 33 dBm of electromagnetic power. This is the maximum power admissible in the spectrometer.

In order to avoid the heating of the sample by the RF pulses we observe the free decay with very small angle pulses (0.2° in solid \(^3\)He at the lowest temperatures). The free precession is damped by the inhomogeneity of the applied magnetic field \( \approx 3 \times 10^{-5} \) with a characteristic time \( T_2^* \) of the order of 40 μs. The signal is displayed on a fast digital transient recorder. The signal to noise ratio is better than 256 in solid \(^3\)He at the lowest temperatures.

- The third parameter is the pressure which is measured at room temperature with a fast response gauge as shown in the insert of figure 2.

![Fig. 1. - Low temperature cell : VJ : vacuum jacket ; S : shield at 0.5 K ; MC : mixing chamber ; CW : copper wires ; SM : superconducting magnet ; C : cell ; HE : heat exchanger ; F : filling capillary ; N : NMR coil ; CR : carbon resistor.](image1)

![Fig. 2. - Melting curve of pure \(^3\)He. The full line refers to the non-polarized \(^3\)He and the dotted line is the calculated melting curve at constant magnetization for polarized \(^3\)He (see ref. \([1]\)). In the insert is sketched the decompression circuit.](image2)
The dashed area between these two curves corresponds to a mixture of polarized solid and liquid.

For practicality, the $^3$He is first compressed up to 50 bar at a temperature of 1.2 K. The solid He is cooled down to 20 mK (point A on figure 2). A plug of solid $^3$He is formed in the filling capillary at a point where the temperature is 0.3 K. This procedure avoids the use of a Pomeranchuk cell and provides a homogeneous temperature in the cell. Then the magnetic field is applied and the decompression is performed by connecting at room temperature the filling capillary to a tank, the volume of which is such that the final pressure of the whole system is 22 bar. A microvalve controls the decrease of the pressure as a function of time.

When the pressure in the capillary is decreased down to 29 bar (the plug is out of the magnetic field and is governed by the usual melting curve) the pressure is released in the cell and after melting the point B is reached; it corresponds to an increase of temperature due to the reverse of the Pomeranchuk effect.

The initial conditions before a decompression are the following: Molar volume of $^3$He = 23.6 cm$^3$/mole $T_i \approx 20$ mK corresponding to a polarization of $H_0 \approx 3.5$ T about 15%.

The $^3$He we used contains about 200 ppm of $^4$He impurities.

3. Experiments and results. — One can classify our experiments in 2 types:

- measurements under static conditions in order to establish the equilibrium properties of the system (measurements of $T_1$ at different pressures and temperatures);
- dynamic measurements of $M$, $P$ and $T$ during decompression.

3.1 Measurements at equilibrium. — In order to define the dynamical properties of the system which are known to vary strongly according to the experimental conditions (geometry, purity of helium, coating of the cell), we have performed measurements of relaxation times in the liquid as well as in the solid, in the cell where the experiment is performed.

The relaxation time in the liquid state is measured by the following procedure: the equilibrium magnetization is first tipped to an angle of about 60° by applying a sequence of 6° pulses of 10 µs length and repetition rate of 2 per s; the recovery of the longitudinal magnetization is observed by applying successive 0.2° pulses every 20 s.

The experimental result for $T_1$ is given in figure 3; the temperature range extends from 50 mK to 1.2 K.

The $T_1$ relaxation time tends to follow a $T^{-2}$ law in the limit of low temperature and presents a minimum of the order of 130 s at 300 mK. Its general behaviour is in agreement with other results [3] and reflects the temperature variation of the spin diffusion coefficient $D$. The magnitude $T_1$ is slightly reduced, probably due to relaxation of $^3$He on the walls and on the copper-wires, which implies $D$ as well.

The values of $T_1$ for the solid state are given in figure 3; above 200 mK they are in agreement with previous results [4]; the high $T$ part corresponds to the contribution of vacancies to the relaxation at our frequency; around 300 mK, we observe the same plateau, with the right value, which is due to the exchange contribution. Below 200 mK, $T_1$ is more difficult to interpret. In fact, at these low temperatures and high fields, the magnetic specific heat of the spins is several orders of magnitude larger than the specific heat of the phonons of the solid. So that once the magnetization has been tilted as a result of a sequence of RF pulses, the lattice cannot restore equilibrium by itself; it rises quickly to the temperature of the Zeeman reservoir and is unable to relax the magnetization; on the contrary, the thermalization process takes place by the coupling with the dilution mixing chamber and it is very long (several minutes) at these temperatures; the lattice is the channel through which the energy flows between the Zeeman reservoir and the dilution mixing chamber.

3.2 Decompression experiments. — The measurement of the magnetization is done by the same procedure used to observe $T_1$, with pulse angles of 0.2°; the repetition rate of the measuring pulse can be increased up to two per second.
4. Discussion of the results. — The main result of these experiments is that the existence of the highly polarized liquid is proved. After decompression the non-equilibrium magnetization is much larger than the magnetization predicted by (1); it can be observed on a time scale $T_1^* = 150 \text{s}$. But several remarks must be added:

1) First of all, the temperature $T_f$ of the final state, about 300 mK in our experiments, corresponds to the minimum of the melting curve. Of course $T_f$ is dependent on the initial conditions: not only of $P_i$ and $T_i$ but also of $M_i$.

2) Once the equilibrium pressure is established, the magnetization decreases with a relaxation time $T_i$ which is the $T_1$ measured at the same temperature (see curve 3). The relaxation of the polarization on a long time scale occurs with a single time constant $7$. This indicates that a unique relaxation process describes the phenomenon and since $T_1^*$ is equal to $T_i$ in the liquid, this supports the idea of a direct transfer of polarization during decompression from the solid at $T_i$ to the liquid at $T_f$.

3) The initial drop of magnetization is the most unexpected observation because it can be as large as 75 % in a slow decompression. A first contribution of the order of 14 % to this drop is simple to explain: when we pass from the highly compressed solid to the liquid at 21 bar, going from 23.6 cm$^3$/mole to 27.3 cm$^3$/mole there is a flow of liquid helium going out of the cell.

But how can we explain 50 % or 75 %? We are then led to admit that the initial decrease of the magnetization with a time constant of 1.8 s is due to a new relaxation process which takes place at the interface between the solid and the liquid. The longer the melting takes, the larger is the effect of this process. But it is not clear from our experiment whether that relaxation process is due to the movement of the melting front, including for instance a fluctuating
local field, or if it is only due to the coexistence of solid and liquid helium. This is clearly a question to be faced directly.

On the other hand, the existence of polarized liquid phase being proved [5], it remains necessary to increase still further the polarization of the initial solid; it would not be difficult in the near future to start from 8 T, 5 mK; the polarization will be as high as 80% and many of the effects predicted by Castaing and Nozières [1] will be enhanced; in particular the melting curve $p(T)$ could be shifted significantly in comparison with the non-polarized case. Many of these effects depend on $M^2$ and were difficult to detect in our first experiment.

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References


See also: Liquid and Solid Helium, J. Wilks (Clarendon Press, Oxford) p. 460.
[5] Evidence for this polarized state has also been found in Copenhagen by Chapellier, M., Frossati, G. and Rasmussen, F. B., using a Pomeranchuk cell (private communication).